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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant(s): Paul G. Clemmer

Application No: 09/816,529

Filing Date: March 23, 2001

Attorney Docket No: 30-4336 (4510)

Title: PURIFICATION OF DIFLUOROMETHANE

Art Group: 1764

Examiner: V. Manoharan

CERTIFICATE OF MAILING

I hereby certify that this correspondence is being deposited with thee Untied States Postal Service with sufficient postage as First Class Mail in an envelope addressed to: Mail Stop AF, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on January 5, 2004.

STEPHEN J. DRISCOLL

Mail Stop AF
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

DECLARATION UNDER 37 C.F.R. §1.131

1. I am the sole inventor in the above-identified patent application.

2. Prior to February 21, 2001, I had completed my invention as described and claimed in the subject application in this country as evidenced by the following:

   a. A draft of the above-identified patent application (herein "draft application") which is an edited verison of an earlier draft I received from Honeywell's outside counsel, Stephen Driscoll. I transmitted this draft via e-mail prior to February 21, 2001 to Honeywell's inside counsel, Colleen Szuch, with instructions that she should forward the draft application to

Mr. Driscoll for finalization. A copy of this draft application as received by Mr. Driscoll has been printed out and attached hereto as Exhibit A, along with the email message received by Mr. Driscoll forwarding the draft application.

b. Schematic drawings which were prepared prior to the draft application and illustrate two preferred embodiments of the invention. These schematic drawings are attached hereto as Exhibit B. It is worthwhile to note that the handwritten reference numbers were added to the figures by others pursuant to preparing the draft patent application.

3. The draft application provides experimental evidence demonstrating that dichloromethane is an effective extractive agent for removing a variety of impurities from HFC-32 using extractive distillation. Specifically, the experimental evidence shows that, in the purification distillation of HFC-32, the level of dichlorofluoromethane, dichlorodifluoromethane and chlorofluoromethane were reduced considerably in the distillate by adding dichloromethane.

4. The draft application and schematic drawings also provide detailed descriptions and illustrations of two preferred embodiments of a fluorination/purification process in which a stream rich in dichloromethane is extracted from a distillation process (which uses dichloromethane as an extraction agent) and is recycled to a fluorination reaction (which uses dichloromethane as a starting material). For example, with respect to one of the embodiments, the draft application states as follows:

In Figure 1, a distillation column 10 is shown having feed lines 1 and 2 connected to its center portion. An overheads flow line 4a leads from the top of the column 10 to a condenser 11. A product flow line 4 leads from the top of the condenser 32 while a reflux flow line 4b leads from the bottom of the condenser back to the distillation column 10. A side flow line 5 leads from the center portion of the column 10 to a fluorination reactor (not shown). A bottoms flow line 6a leads from the bottom of the column 1 to a reboiler 12 with a vapor return line 6b leading from the top of the reboiler back to the bottom of the column 10. A flow

line 6 leads from the bottom of the reboiler 12 and is fed to the center portion of the column 10 as recycle feed line 3. A diversion valve 13 is disposed along flow line 6 to divert a desired portion of the flow to flow line 7 which optionally connects to a fluorination reactor (not shown).

(Draft application, p.8). Although the draft application does not provide experimental data on the particular embodiments described therein, their efficacy is obvious in light of their description and schematic illustrations. Therefore, the draft application supports my assertion that I not only conceived of the invention prior to February 21, 2001, but also reduced it to practice before that date.

5. Any dates deleted from Exhibits A or B were prior to February 21, 2001.

6. As the person signing below:

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful statements may jeopardize the validity of the application or any patent issued thereon.

Jan. 2, 2004
Date

Paul G. Clemmer
Paul G. Clemmer

M:\SDriscoll\Honeywell-(Allied)\22972\DECLARATION UNDER 37 C.F.R. §1.131.wpd

PURIFICATION OF DIFLUOROMETHANE

FIELD OF INVENTION

The present invention relates to the purification of difluoromethane (HFC-32). More specifically, this invention relates to a method and system for separating HFC-32 from an azeotropic or near-azeotropic mixture.

BACKGROUND OF THE INVENTION

Hydrofluorocarbons (HFCs) have been identified as commercially-viable substitutes for chlorofluorocarbons in various applications. For example, difluoromethane (HFC-32) is useful as a refrigerant, blowing agent, cleaning agent, and aerosol propellant just to name a few applications. Such HFCs are prepared commonly by fluorinating chlorinated organic starting materials, such as dichloromethane, using a fluorination agent, such as hydrogen fluoride. Although fluorination has proved to be a convenient method for preparing HFCs, particularly HFC-32, the introduction of chlorinated impurities in the product stream is generally unavoidable. Such chlorinated impurities include, for example, starting materials and intermediates from incomplete fluorination and/or by-products from undesirable side reactions. These chlorinated impurities diminish HFC-32's purity.

Although significant purification of HFC-32 can be achieved using conventional distillation, certain chlorinated impurities such as dichlorodifluoromethane (CFC-12) and methyl chloride (HCC-40) tend to form azeotropic or near-azeotropic mixtures with HFC-32 thereby rendering conventional distillation impossible or impractical. Generally, if the relative volatility of the product to the impurities in the mixture close to 1.0, conventional distillation is not practical. The relative volatility α of an impurity 2 to a

fluorinated compound 1 in a mixture is defined herein according to the following equation:

$$\alpha_{2,1} = \frac{y_2}{x_2} \times \frac{x_1}{y_1}$$

where x and y are are, respectively, the liquid and vapor mole fraction of the components.

Therefore, for a mixture of HFC-32 and a chlorinated impurity, such as CFC-12 and/or HCC-40, where the relative volatility is close to 1.0, a separation technique other than conventional distillation is desirable.

Alternatives to conventional distillation for separating azeotropic or near-azeotropic mixtures include adsorption, membrane diffusion, and extractive distillation. Perhaps the most popular of these approaches is extractive distillation. For example, in Japanese Patent Publication No.7-291878, an extractive distillation technique is disclosed in which an extractive agent selected from 1,1-dichloro-1-fluoroethane, 2,2-dichloro-1,1,1-fluoroethane, trichlorotrifluoroethane, and dichloropentafluoropropane is used in separate 1,1,1-trifluoroethane, pentafluoroethane or methyl chloride from HFC-32. Unfortunately, despite their effectiveness, the extractive agents described above nevertheless add impurities to the process that must be removed through additional distillation steps. These additional distillation steps add cost and complexity to the process.

Therefore, a need exists for a process of purifying HFC-32 without adding extractive agents that complicate the process and necessitate additional distillation steps. The present invention fulfills this need among others.

DESCRIPTION OF THE INVENTION AND PREFERRED EMBODIMENTS

The present invention provides both for the purification of HFC-32 by extraction distillation using dichloromethane, and for a unique fluorination/distillation configuration which avoids the need for additional distillation steps.

It has been found, surprisingly, that dichloromethane forms a non-ideal liquid-vapor mixture with HFC-32 and thus is suitable as an extraction agent in the extractive distillation of HFC-32 from a mixture of impurities. More specifically, the mixture of HFC-32 and dichloromethane exhibits non-ideal behavior according to Raoult's Law such that an increase in the concentration of dichloromethane results in a less-than-expected reduction in the mixture's volatility. Dichloromethane does not exhibit, however, the same non-ideal behavior with impurities typically found in the preparation of HFC-32. Therefore, an increase in the relative volatility between HFC-32 and the impurities can be effected by the addition of dichloromethane. The increase in relative volatility in turn provides for more effective and efficient distillation.

Accordingly, one aspect of the present invention is a method for separating difluoromethane from a mixture of the difluoromethane and at least one impurity using dichloromethane as an extractive agent. In a preferred embodiment, the method comprises extractively distilling mixture using dichloromethane as the extractive agent to recover purified difluoromethane having a concentration of the impurity lower than that of the mixture.

Aside from forming a non-ideal mixture with HFC-32, dichloromethane is particularly advantageous as an extractive agent since it also is a starting material in the fluorination of HFC-32. Consequently, after effecting increased relative volatility between the constituents of the reactor stream and thereby facilitating extractive distillation, the dichloromethane and impurities may be removed from the distillation unit

and used to feed the fluorination reaction. Thus, the need for additional distillation steps to separate the extractive agent from the impurities is eliminated.

The general concept of supplying the fluorination reaction with a mixture of the extractive agent and removed impurities is disclosed in U.S. Patent No. 5,200,431. As disclosed therein, a bottoms stream comprising the extractive agent, trichloroethane, and impurities are fed to the fluorination reactor. Although this approach avoids the need for additional distillation steps, it makes the distillation and fluorination operations interdependent upon one another. This interdependency generally requires that the amount of extractive agent used in the extractive distillation process be no more than that consumed in the fluorination reaction. Otherwise, the extractive agent/starting material will build up in the fluorination reactor thereby reducing productivity.

The present invention overcomes this interdependency problem by providing a novel fluorination/distillation configuration that allows for the independent operation of the fluorination and extractive distillation processes. More specifically, the extractive distillation is performed such that the extractive agent is fractionalized into a side stream and a bottoms stream. The side stream comprises a mixture of the extractive agent and impurities while the bottoms stream has a relatively-low concentration of impurities. The side stream may be supplied to the fluorination reaction, while the bottoms stream is sufficiently low in impurities that it can be recycled into the distillation column. This configuration enables the amount of extractive agent recycled to the fluorination reaction and to the extractive distillation column to be adjusted for optimum performance. More specifically, the flow rates of the side stream and bottoms stream may be adjusted to optimize fluorination and distillation conditions. It is worthwhile to note that this configuration is not limited to HFC-32, but can be used wherever the extractive agent is also a starting material or an intermediate of the fluorinated product.

Accordingly, another aspect of the present invention is a method of preparing a fluorinated compound where extractive distillation supplies the fluorination reaction with a controllable feed recycle stream. In a preferred embodiment, the method comprises: (a) fluorinating a chlorinated organic compound to produce a reactor stream comprising a fluorinated compound and at least one chlorinated impurity; (b) feeding the mixture to a distillation unit; (c) feeding an extractive agent to the distillation unit, wherein the extractive agent is the chlorinated organic compound; (d) operating the distillation unit under conditions sufficient to distill a product stream of the fluorinated compound containing a concentration of the impurity less than that of the reactor stream; (e) withdrawing a side stream from the distillation unit, the side stream comprising the impurity and the extractive agent; (f) withdrawing a bottom stream from the distillation unit, the bottom stream comprising the extractive agent and having a concentration of impurity less than that found in the side stream; (g) supplying the fluorination reaction with at least a portion of the side stream; and (h) recycling at least a portion of the bottom stream into the distillation unit.

Yet another aspect of the invention is a system for preparing a fluorinated compound which provides for independent recycle streams to the fluorination and distillation operations. In a preferred embodiment, the method comprises: (a) a reactor adapted to receive a chlorinated organic compound from at least one source and a fluorination agent for fluorinating the chlorinated organic compound, the reactor adapted to facilitate fluorination of the chlorinated organic compound to produce a reactor stream comprising a mixture of a fluorinated compound and at least one chlorinated impurity; (b) a first conduit for feeding the reactor stream to a distillation unit; (c) a distillation unit configured to receive the mixture and an extraction agent from at least one source, the distillation unit being adapted to facilitate extractive distillation of the mixture to produce an overheads stream, a side stream, and a bottoms stream, the overheads stream

comprising the fluorinated compound and a lower concentration of chlorinated impurities than that of the reactor stream, the side stream comprising the extractive agent and the chlorinated impurity, the bottoms stream comprising the extractive agent and a concentration of the chlorinated impurity less than that of the side stream; (d) a second conduit for supplying at least a portion of the side stream to the fluorination reactor; and (e) a third conduit for recycling at least a portion of the bottoms stream to the distillation unit.

Considering now the purification of HFC-32 in more detail, the invention is applicable to any mixture of HFC-32 and one or more impurities having a volatility above that of dichloromethane. The purification method is especially applicable to mixtures obtained in the preparation of HFC-32 through the fluorination of chlorinated organic compounds. Such mixtures usually comprise HFC-32, hydrogen fluoride, and at least one chlorinated impurity (usually a starting material, intermediate, or unwanted byproduct) having the formula:

$$CH_wCl_y X_z$$

wherein each X is an independently selected halogen, $y \geq 1$ and $w+y+z=4$. Typically, X is fluorine. For example, chlorinated impurities typically found in a reactor stream of HFC-32 include chlorofluoromethane (HCFC-31), chloromethane (HCC-40), dichloromethane (HCC-30), chlorodifluoromethane (HCFC-22), chlorotrifluoromethane (CFC-13), dichlorodifluoromethane (CFC-12) and combinations of two or more thereof. The method is particularly appropriate for azeotropic or near-azeotropic mixtures, such as mixtures of HFC-32 and CFC-12 and/or HCC-40. It is worthwhile to note, however, that the method is not restricted to azeotropic or near-azeotropic mixtures and can improve the distillation of various mixtures by increasing the relative volatility between the constituents.

The concentration of the chlorinated impurity in the reactor stream being treated in accordance with the invention is typically from 20 to 5000 ppm by weight, but mixtures containing smaller or larger amounts of the chlorinated impurity also may be separated.

If desired, the reactor stream may be pre-treated to effect partial or essentially complete removal of one or more other chlorinated impurities and/or hydrogen fluoride before performing the separation method of the present invention. Such a pretreatment step is preferable where the relative volatility of the product and an impurity is greater than about 1.1 such that conventional distillation is readily achieved without the use of a particularly tall distillation column. Furthermore, some form of pretreatment may be preferred if the impurity has a lower volatility than dichloromethane in which case combining it with dichloromethane would diminish the relative volatility of the mixture.

The method of the invention may be performed using conventional extractive distillation procedures. The distillation preferably is conducted to effect a distillation stream of purified HFC-32 having less than about 50 ppm (by weight) chlorinated impurities. More preferably, the reaction is conducted to effect a distillation stream having less than about 10 ppm chlorinated impurities.

In accordance with the present invention, high yields of HFC-32 are realized. In a preferred embodiment, the yield of HFC-32 is no less than about 80% and, more preferably, no less than about 90%.

Generally, it is preferable to conduct the extractive distillation near or above atmospheric pressure to minimize operation costs. For example, suitable results have been obtained operating at a pressure of about 1 to about 15 bars. The temperature at which the distillation unit is operated depends upon the operating pressure.

It is particularly advantageous that dichloromethane is both an extractive agent and a starting material in the preparation of HFC-32. This way, the bottoms fraction from the extractive distillation unit comprising dichloromethane and other chlorinated species can be supplied to the fluorination reaction. In a preferred embodiment of the present invention, rather than feeding the bottoms fraction to the fluorination reaction, a configuration is used which fractionalizes the extractive agent into two streams: a side stream comprising extractive agent mixed with impurities and a bottoms stream comprising extractive agent and having a low concentration impurity. The bottoms stream preferably has no greater than about 5000 ppm (by weight) impurities, and, more preferably, no greater than about 1000 ppm. The side stream is supplied to the fluorination reaction and the bottoms stream is recycled to the distillation unit. Thus, the amount of feed supplied to the fluorination reactor can be reduced by increasing the amount of the bottoms stream recycled to the distillation unit and vice versa.

Referring now to the drawings, two preferred embodiments of the system are schematically illustrated. In Figure 1, a distillation column 10 is shown having feed lines 1 and 2 connected to its center portion. An overheads flow line 4a leads from the top of the column 10 to a condenser 11. A product flow line 4 leads from the top of the condenser 32 while a reflux flow line 4b leads from the bottom of the condenser back to the distillation column 10. A side flow line 5 leads from the center portion of the column 10 to a fluorination reactor (not shown). A bottoms flow line 6a leads from the bottom of the column 1 to a reboiler 12 with a vapor return line 6b leading from the top of the reboiler back to the bottom of the column 10. A flow line 6 leads from the bottom of the reboiler 12 and is fed to the center portion of the column 10 as recycle feed line 3. A diversion valve 13 is disposed along flow line 6 to divert a desired portion of the flow to flow line 7 which optionally connects to a fluorination reactor (not shown). In operation, an impure reactor stream containing a fluorinated compound, such as HFC-32,

and one or more chlorinated impurities is fed into a distillation column 10 through feed line 1 along with an extractive agent, for example, dichloromethane, through feed line 2. Optionally, a recycle stream also may be fed to the column via the recycle feed line 3. The distillation column 10 is operated at conditions and with reflux ratios sufficient to effect distillation of a purified fluorinated compound through a product stream in the product flow line 4. The product stream comprises the fluorinated compound and a concentration of impurity less than that of the reactor stream. Removal of impurities from the distillation unit 10 is effected through a side stream through side flow line 5. The side stream optionally may be fed to the fluorination reactor (not shown). The bottoms stream leaving through the bottoms flow line 6 is rich in extractive agent and has a concentration of impurities less than that of the side stream. Optionally, it may be recycled back to the distillation column 10. According to this configuration, the distillation unit not only provides for the separation of the impurity from the product but also provides for the separation of extractive agent from a mixture of extractive agent and one or more impurities. This way, the impurities are removed from the distillation unit but a recycle stream is still available. The location of the feed points, temperature and relative flow rates of the feed streams, reflux ratios, column size and distillation operating parameters are adjusted to optimize the process operation to achieve the desired separation. Although the use of this configuration has been described with respect to dichloromethane, this configuration can be practiced wherever the extractive agent is a starting material or intermediate of the fluorination reaction.

In Figure 2, an alternative distillation system is shown schematically. A distillation column 20 is shown with a feed line 21, an extractive agent feed line 22, and a recycle feed line 23 connected to its center portion. An overheads flow line 24a leads from the top of the column 20 to a condenser 30. A product flow line 24a leads from the top of condenser 30 while reflux flow line 24b leads from the bottom of the condenser 30

back to the top of the column 20. A bottoms flow line 26a leads from the bottom of the column 20 to a reboiler 31. A vapor return line 26b leads from the top of the reboiler back to the bottom of the column 20 and a flow line 26 leads from the reboiler 31 to a second distillation column 27. An overheads flow line 25a leads from the top of the column 27 to a condenser 28. An impurity flow line 25 leads from the top of condenser 28. A reflux flow line 25b leads from the impurity flow line 25 back to the top of the column 20. A bottoms flow line 23a leads from the bottom of the column 27 to a reboiler 29. A vapor return line 23b leads from the top of the reboiler 29 back to the bottom of the column, while an extractive agent flow line 23 leads from the bottom of the reboiler 29 and optionally feeds the distillation column 20.

In operation, the impure reactor stream is fed through feed line 21 and the extractive agent, for example, dichloromethane is fed through the feed line 22 to the first distillation column 20. Optionally, a recycle stream also may be fed to the first distillation column through recycle feed line 23. The distillation column 20 is operated at conditions sufficient to effect distillation of a purified fluorinated compound, such as HFC-32, through a product stream in the product flow line 24. Removal of impurities and extractive agent is effected through a bottoms stream in line 26. This bottoms stream then is fed to the second distillation unit 27. Impurities are removed as distillate in an impurities stream through line 25, while extractive agent is removed in a bottoms stream through line 23. The bottoms stream, rich in extractive agent, optionally may be recycled to the first distillation unit 20. Again, the location of the feed points, temperature and relative flow rates of the feed streams, column size and distillation operating parameters are adjusted to optimize the process operation to achieve the desired separation.

In both configurations described above, to lower operating costs, all columns in the scheme preferably operate at near- or super-atmospheric pressure. The recycle streams preferably are cooled before being fed to the distillation units. Furthermore, the

product obtained by the method of the invention may be subjected, as desired, to further purification procedures.

Example:

A sample of impure HFC-32 produced by fluorination of dichloromethane was fed to a stainless steel distillation column with approximately 40 theoretical stages equipped with a reboiler and condenser, operating at a pressure of approximately 11.5 bar. Table 1 gives the amount of several impurities as a ratio to the HFC-32 content, as measured in the feed and at the top and bottom of the distillation column. The results demonstrate that the impurities were not effectively removed by conventional distillation.

Table 1

Impurity	Feed liquid (wt%)	Distillate (wt%)	Reboiler vapor (wt%)
Methyl chloride	0.0039	0.0010	0.0060
Dichlorofluoromethane	0.1430	0.0436	0.1659
Dichlorodifluoromethane	0.0876	0.1626	0.0232
Chlorofluoromethane	0.6923	0.0003	1.04

The feed of impure HFC-32 was then continued at approximately the same distillation conditions to test the feasibility of removing impurities from HFC-32 by extractive distillation using dichloromethane as the extractive agent. Dichloromethane was fed to the column at a location above the HFC-32 feed point at a mass feed rate 6.5 times that of

the HFC-32 feed. Table 2 gives the amount of several impurities as a ratio to the HFC-32 content, as measured in the feed and at the top and bottom of the distillation column while doing this extractive distillation. These results demonstrate that dichloromethane is an effective extractive agent for removing the listed impurities from HFC-32 by extractive distillation.

Table 2

Component	Feed liquid (wt%)	Distillate (wt%)	Reboiler vapor (wt%)
Methyl chloride	0.0039	0.0002	0.0111
Dichlorofluoromethane	0.1430	0.0146	0.3660
Dichlorodifluoromethane	0.0876	0.0053	0.0870
Chlorofluoromethane	0.6923	0.0002	1.342

CLAIMS

WHAT IS CLAIMED IS

1. A method for separating difluoromethane from a mixture of said difluoromethane and at least one impurity, said method comprising extractively distilling said mixture using dichloromethane as the extractive agent to recover a product stream of purified difluoromethane having a concentration of said impurity lower than that of said mixture.

2. The method of claim 1, wherein the step of extractively distilling produces at least one extractive agent stream comprising a mixture of said impurity and dichloromethane; and wherein said process further comprises:

supplying at least a portion of said extractive agent stream to a fluorination reaction which produces said difluoromethane.

3. The method of claim 1, wherein the step of extractively distilling produces a side stream and a bottoms stream, said side stream comprising a mixture of dichloromethane and said impurity, said bottoms stream comprising dichloromethane and a concentration of said impurity less than that of said side stream; and wherein said method further comprises:

supplying at least a portion of said stream to a fluorination reaction which produces said difluoromethane.

4. The method of claim 1, wherein said impurity is a chlorinated impurity having the formula:

$CH_wCl_y X_z$

wherein each X is an independently selected halogen, $y \geq 1$ and $w+y+z=4$.

5. The method of claim 4, wherein X is fluorine.

6. The method of claim 4, wherein said chlorinated impurity is selected from the group consisting of chlorofluoromethane, chloromethane, chlorodifluoromethane, dichlorodifluoromethane and combinations of two or more thereof.

7. The method of claim 6, wherein said chlorinated impurity is selected from the group consisting of dichlorodifluoromethane, chloromethane and combinations thereof.

8. The method of claim 1, wherein the concentration of impurity in the recovered HFC-32 is no greater than about 50 ppm (by weight).

9. The method of claim 8, wherein the concentration of impurity in the recovered HFC-32 is no greater than about 10 ppm.

10. The method of claim 1, wherein the yield of HFC-32 is no less than about 80%.

11. A method as claimed in claim 1, wherein the step of extractively distilling is conducted at a pressure of about 1 to about 15 bars.

12. A method for preparing a fluorinated compound, said fluorinated compound being produced by the fluorination of a chlorinated organic compound, said method comprising:

fluorinating a chlorinated organic compound to produce a reactor stream comprising a mixture of a fluorinated compound and at least one impurity;

feeding said mixture to a distillation unit;

feeding an extractive agent to said distillation unit, wherein said extractive agent is said chlorinated organic compound;

operating said distillation unit under conditions sufficient to distill a product stream comprising said fluorinated compound and a concentration of said impurity less than that of said reactor stream;

withdrawing a side stream from said distillation unit, said side stream comprising said impurity and said extractive agent;

withdrawing a bottom stream from said distillation unit, said bottom stream comprising said extractive agent and a concentration of said impurity less than that of said side stream;

supplying the fluorination reaction with at least a portion of said side stream; and

recycling at least a portion of said bottom stream to said distillation unit.

13. The method of claim 12, wherein said extractive agent is selected from the group consisting of dichloromethane, trichloroethylene, tetrachloroethylene, and 1,1,1,3,3-pentachloropropane.

14. The method of claim 12, where said fluorinated compound is selected from the group consisting of difluoromethane, 1,1,1,2-tetrafluoroethane, pentafluoroethane, and 1,1,1,3,3-pentafluoropropane.

15. The method of claim 12, wherein said extractive agent is dichloromethane and said fluorinated compound is difluoromethane.

16. A method as claimed in claim 12 wherein the step of extractively distilling is conducted at a pressure of about 1 to 15 bars.

17. A system for the preparation of a fluorinated compound comprising:

a reactor adapted to receive a chlorinated organic compound from at least one source and a fluorination agent for fluorinating said chlorinated organic compound, said reactor adapted to facilitate fluorination of said chlorinated organic compound and to produce a reactor stream comprising a mixture of a fluorinated compound and an impurity;

a first conduit for feeding said mixture to a distillation unit;

a distillation unit configured to receive said mixture and an extraction agent from at least one source, said distillation unit being adapted to facilitate

extractive distillation of said mixture to produce an overheads stream, a side stream, and a bottoms stream, said overheads stream comprising said fluorinated compound having a lower concentration of chlorinated impurities than said reactor stream, said side stream comprising said extractive agent and said chlorinated impurity, said bottoms stream comprising said extractive agent and having a lower concentration of said chlorinated impurity than said side stream;

a second conduit for supplying at least a portion of said side stream to said fluorination reactor; and

a third conduit for supplying at least a portion of said bottoms stream to said distillation unit.

18 The system of claim 16, further comprising:

a device for adjusting the flow rates through said first conduit, said second conduits or a combination thereof.

19 The system of claim 17, wherein said device is a valve in said third conduit for diverting a portion of the bottoms stream from said distillation unit to said fluorination reactor.

ABSTRACT

A method for separating difluoromethane from a mixture of said difluoromethane and at least one impurity, said method comprising extractively distilling said mixture using dichloromethane as the extractive agent to recover a product stream of purified difluoromethane having a concentration of said impurity lower than that of said mixture.

Stephen J. Driscoll

From: Szuch, Colleen D.
Sent:
To: Sdriscoll@synnlech.com
Cc: Clemmer, Paul G.
Subject: FW: 30-4336 draft


22972.dr1


22972mod.dr1.doc

FYI and action.

Chief Intellectual Property Counsel
Specialty Chemicals
HONEYWELL
973 455 - 2857 phone
973 455 - 5817 fax
colleen.szuch@honeywell.com

-----Original Message-----
From: Clemmer, Paul G.
Sent:
To: Szuch, Colleen D.
Subject: Re: 30-4336 draft

Colleen,
I finished my revisions of the patent application draft on File # 30-4336 prepared by Steve Driscoll. It's been so long that I don't know if Steve is still working on this case, or if his e-mail address has changed (he didn't respond to my e-mail and I don't have his phone #). So, I'm sending you my version (22972mod.dr1.doc) and also attaching Steve's original version (22872.dr1).

Paul Clemmer

-----Original Message-----
From: Clemmer, Paul G.
Sent:
To: 'Steve Driscoll'
Subject: RE: S&L 22,972 Purification of Difluoromethane

I have (finally) been able to spend some time on this. I finished my review of your draft on this case and prepared a draft based on my revisions. If you are still involved with this case, let me know so I can send you my revision.

Thanks,
Paul Clemmer

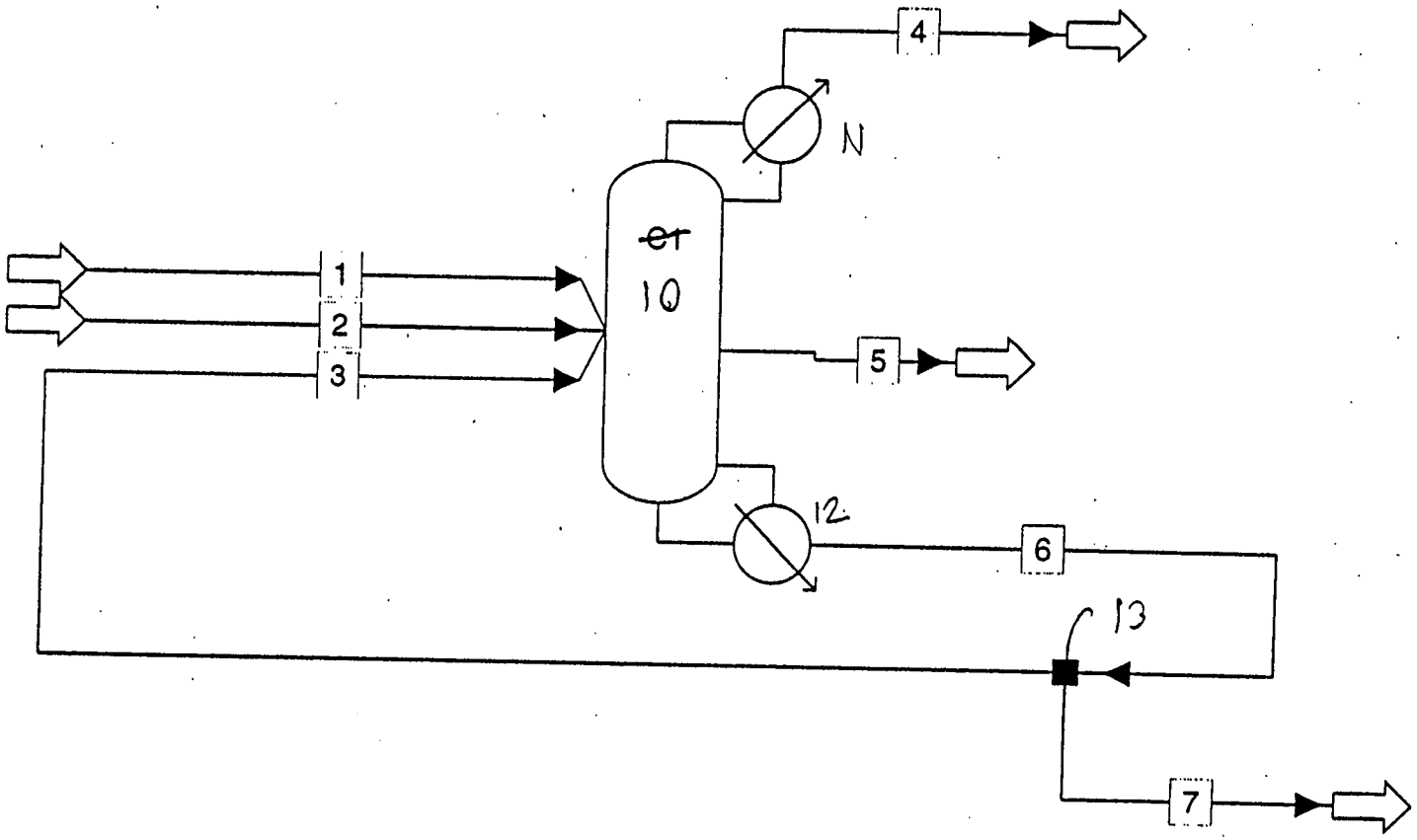
-----Original Message-----
From: Steve Driscoll [mailto:SDriscoll@svnnlech.com]
Sent:
To: Clemmer, Paul G.
Subject: Re: S&L 22,972 Purification of Difluoromethane

How is your review of the application coming along? We look forward to receiving your comments soon so that we can file the application before

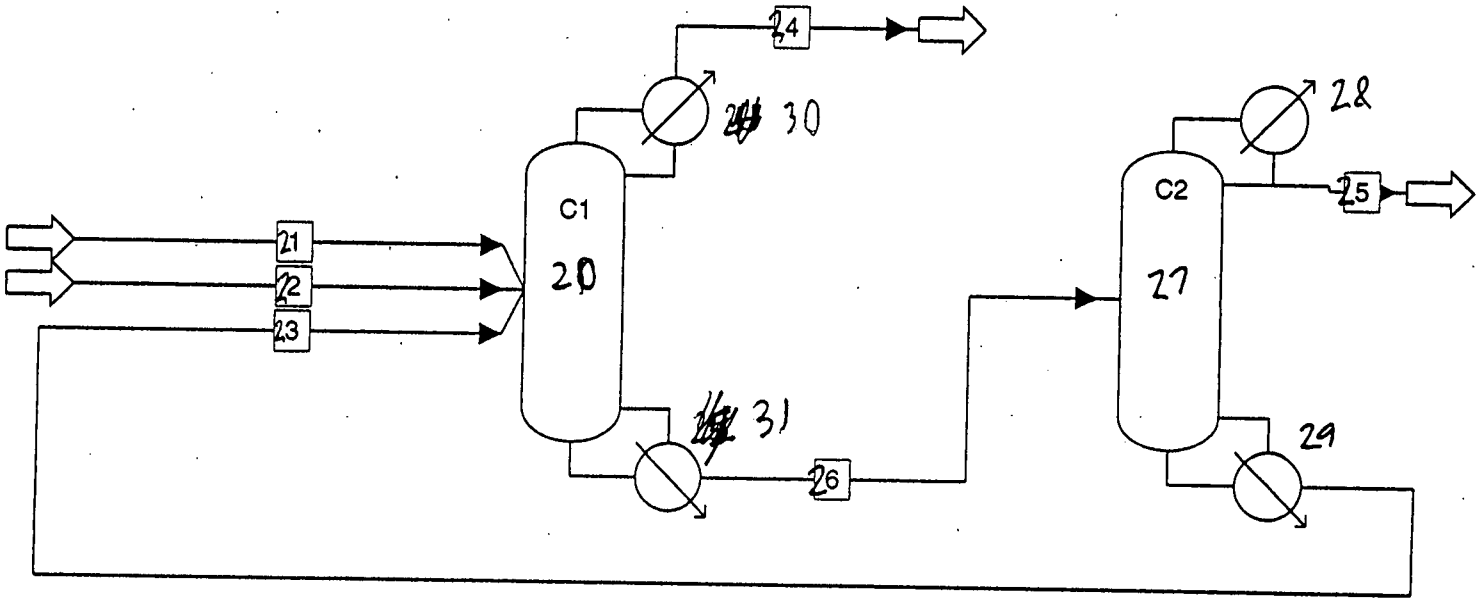
Best regards,
Steve

Clemmer, Paul G. wrote:

> Sorry I haven't gotten back to you sooner. I have reviewed the draft, and I
> think we need to include additional information from our lab tests. I'll
> send my comments and copies of the additional documents as soon as I can.
>
> Paul
>
> -----Original Message-----
> From: Steve Driscoll [mailto:SDriscoll@synnlech.com]
> Sent:
> To: paul.clemmer@alliedsignal.com
> Subject: S&L 22,972 Purification of Difluoromethane
>
> Dear Paul:
>
> Have you had the opportunity to review the draft of the above-referenced
> application transmitted to you on
>
> Please provide me with your comments to the above-referenced draft asap.
> Thank you.
>
> Best regards,
> Steve



DRAWING 1



DRAWING 2